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Prospect of Molecular Sieves Production using Rice Husk in Bangladesh: A Review

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Abstract— *A growing need of molecular sieves, now a day, in many industries of Bangladesh makes the materials most important and attracted special attention. The review explores each step involved in the whole process of molecular sieves preparation from rice husk (RH). Many refineries and chemical industries are using molecular sieves as adsorbent material and process catalyst very efficiently. Rice husk is one of the potential sources of biomass to get a maximum percent of* SiO_2 *that is required to synthesize molecular sieves. 93.2% of* SiO_2 *can be extracted from rice husk ash (RHA). In the fiscal year of 2016-2017, about 6.86 million tons of rice husk are produced in Bangladesh whereas worldwide production of rice husk is approximately 97.24 million tons. Geographical locations, combustion methods, and temperature are influencing factor mainly to get the content of silica and structure of molecular sieves.) Rice husk ash containing amorphous silica is produced at temperatures lower than* 800^oC *(500-*800^oC*). The chemical method with washing pretreatment and post heat treatment are considered as the most simple and successful* SiO_2 *extraction methods from RH. Different types of molecular sieves; 13x, 3A, 4A & 5A can successfully be synthesized via the widely used hydrothermal method. Molar ratio of ingredients, reaction temperature, time, acid-base media, ageing etc. are key factors to be considered in designing different molecular sieves.*

Keywords— ***Molecular sieves, Biomass, Rice husk, Bio-silica, pyrolysis.***

I. INTRODUCTION

A breakthrough and unique opportunity is awaiting for molecular sieves to contribute to nano-technology for various microelectronic devices, adsorbents, corrosion resistance, membranes, Sensors, thermo-electronics, ionic conductors, superconductors and catalysts. Advancement of this technology adds a new dimension to designing nano-porous materials and manipulating numerous physio-chemical properties of molecular sieves[1-4].

Molecular sieves (MS) are absorbent materials having a solid framework and molecular scale crystalline structure like sponges[5]. They have precise pores and cavities interconnected by pore openings across the molecular structure[6]. The main composition of molecular sieves is silicon (Si), aluminum (Al) and oxygen (O)[7]. The crystalline structure of molecular sieves depends on Si/Al ratio, precursors used and preparation conditions[8]. The core features of these pores and cavities are the absorption of gaseous molecules of certain properties while rejects others [9,10]. They are widely used in petroleum oil refinery, petrochemistry, organic synthesis in the chemicals production [11],heavy metal absorption from liquids [12-15], gas refinery [16] etc. Molecular sieves with desired characters can be synthesized from the chemical reaction between silica and alumina compounds [17,18]. But it's not cost efficient norecofriendly. In this situation,silica-rich alternatives are highly important and rice husk(RH) as silica source is widely suggested by authors [19,20]. Many industries not only in Bangladesh but also all over the world have been using molecular sieves for their need. A total collective method of molecular sieves production is very important to know for better utilization of it that discussed in seldom.

The principal focus of the review is to explore molecular sieves production and uses. It offers brief discussion on factors affecting the production process, prospect of rice husk biomass as feedstock in Bangladesh and the methods to obtain the optimum composition of molecular sieves.

II. POTENTIALITY OF RICE HUSK IN BANGLADESH

Agriculture is the basis of the economy of Bangladesh and paddy rice is the major agricultural crop of the country[21]. Bangladesh holds the fourth position in worldwide paddy rice production and the country produced an approximately 33.80 million tons of rice in the fiscal year of 2016-17 [22]. The paddy rice cultivation and processing produces a significant amount of biomass byproduct in the form of straw, rice husk (RH) and rice bran[21]. Generally, straw and bran are used as animal feeding. On the other hand, RH is a waste product that has limited use in power generation sectors of the country. Bangladesh experiences a cumulative increase in paddy rice production. Consequently, the RH production in the country is also increasing. Fig. 1presents the country’s RH production from the fiscal year2011-12 to2016-17[22]. To get the optimum rice husk production, an average of 20 kg husk generation per 100 kg of paddy rice production is considered [23].Hence, approximately 6.86 million tons of RH are produced in the country in the fiscal year of 2016-17.

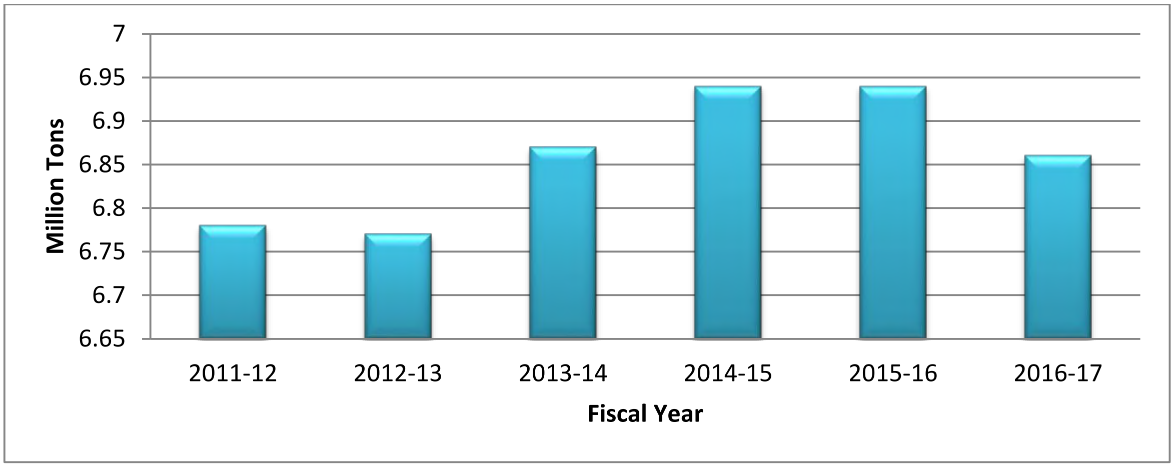

Fig. 1: Rice husk production scenario in recent years in Bangladesh

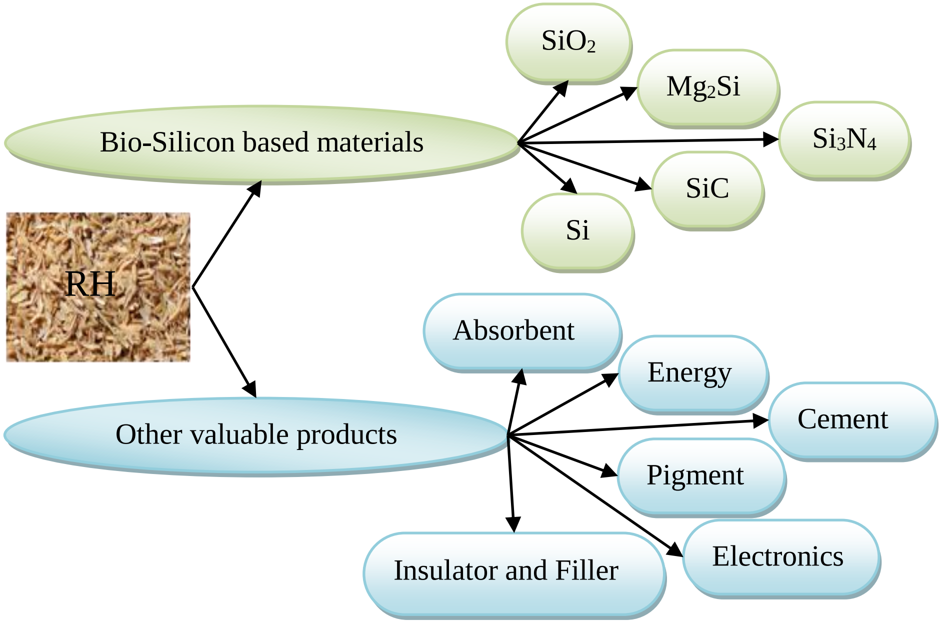

Fig.2: Various possible products from RH.

This mountainous amount of rice husk is poorly managed in the country [24]. A large portion of this huge amount of rice husk is burnt in open for heating, parboiling or cooking purpose, and another portion is dumped for landfilling. Both of these practices are adversely affecting the environment and human health. However, by adopting advanced technologies and taking suitable measures, RH can be used for cleaner energy generation (e.g. bio-oil, product gas, H_2generation etc. by pyrolysis/gasification techniques)and various value-addedproducts such asbio-silica. Fig. 2 shows thevalue-added materials that can be produced from RH[25]. RH derived material especially bio-silica (SiO_2) is very important raw material for various industries like molecular sieve, zeolite, electronics, cement, absorbent and polymer industries etc. RH has higher bio-silica content than other common agricultural byproducts. Thus, RH can be converted into valuable products and it can contribute to our economy rather than causing environmental and health problems.

III. SOURCES OF BIO-SILICA

In the earth crust, silicon (Si) is the second most abundant material after oxygen. Naturally, plants get silicon from the soil as silicic acid (H_4SiO_4). Plants absorb H_4SiO_4 by roots when the P^H of the soil is under 9 [26]. Silicon bonds in H_4SiO_4 are relocated from roots to shoots and deposited as SiO_2 (Opel), which is amorphous in nature. Different plant species exhibit silica accumulations in the shoot ranging from 0.1% to 10% [26,27]. In plants, silicon is accumulated in the form of phytoliths (amorphous hydrated silica) [28].Silicon compound, generally immobile, has a higher concentration in older plant tissues and its accumulation is not consistent among tissues. Guntzer et al. [29] found that 7 out of 10 mostly harvested crops are Si accumulators. Among these high Si accumulating crops rice, sugar cane and wheat have greater silicon withdrawal from the soil than other natural systems. For example, rice and sugarcane have silicon withdrawal rates of 300 kg/ ha/ year and 500 kg/ha/year respectively, where US grasslands withdraw only between 22 to 67 kg/ ha/ year [30-32]. So plants especially crop residues from high silicon accumulating crops can be considered as a good source for bio-silica. Table 1 shows the chemical composition of biomass or crop byproducts highlighting the silica content. Where ash from rice husk (RHA) contains maximum 93.2% silica according to Chindaprasirt et al. [33] followed by ash from bamboo leaf, wheat straw and sugar cane bagasse [34-36]. Other byproducts have less than 70% silica in the ash where paper mill sludge is sitting at the bottom with 25.7% silica in its ash [37].

Table 1: The chemical composition of biomass or crop byproducts after combustion

Biomass residue	SiO_2%	Al_2O_3 %	Fe_2O_3%	CaO%	MgO%	Na_2O%	K_2O%	Reference
Rice husk	93.2	0.4	0.1	1.1	0.1	0.1	1.3	[33]
Bamboo leaf	75.90	4.13	1.22	7.47	1.85	0.21	5.62	[34]
Wheat straw	73	3.90	1.75	8.12	2.80	-	-	[35]
Sugarcane bagasse	72.74	5.26	3.92	7.99	2.78	0.84	3.47	[36]
Sawdust	67.20	4.09	2.26	9.98	5.80	0.08	0.11	[38]
Corn cob	66.38	7.48	4.44	11.57	2.06	0.41	4.92	[39]
Oil palm shell	63.6	1.6	1.4	7.6	3.9	0.1	6.9	[33]
Sugar cane straw	59.06	4.75	3.18	19.59	2.25	0.73	4.75	[36]
Vetiver grass	57.48	3.73	1.71	5.45	1.24	0.12	15.49	[40]
Sewage sludge	50.6	12.8	7.21	1.93	1.48	0.32	1.70	[41]
Paper mill sludge	25.70	18.86	0.87	43.51	5.15	1.56	1.31	[37]

For silica content determination, ash from residues and byproducts are first produced and to do these, researchers occupied different methods. Table 2 presents the ash production methods followed by researchers to get the chemical composition of biomass residues mentioned in Table 1. Correspondingly, rice husk, bamboo leaf, sawdust, corn cob, and sugarcane straw are open-air burnt with or without grinding. Whereas, the ash of sugar cane bagasse and oil palm shell is collected from boilers and power plants. Again, for wheat straw and bamboo leaf furnace is used [34,35]. On the other hand, combustion temperature varies from 600^0C to 900 ^{0}C and the combustion time is maximum of 9 hours for vetiver grass and minimum 1 hour for sugarcane straw.

Table 2: Ash generation methods from biomass residues

Biomass residue	Ash production methods	Ignition temperature, ^{0}C	Ignition time, h	Reference
Rice husk	open burning, ball mill grinding	650	-	[33]
Bamboo leaf	open-air burnt then furnace heated	600	2	[34]
Wheat straw	cut and burned in an electrical furnace then ground	670	5	[35]
Sugarcane bagasse	Extracted directly from boilers of the sugar factory	-	-	[36]
Sawdust	open-air burn and ground	-	-	[38]
Corn cob	Ground and open-air burnt	650	8	[39]
Oil palm shell	Extracted from thermal power plant then ball mill ground	-	-	[33]
Sugarcane straw	open-air burn	800	1	[36]
Vetiver grass	Dried, burnt in ferrocement incinerator and ground	900	9	[40]
Sewage sludge	modular incinerator and ball mill grinding	700	3	[41]

IV. RICE HUSK DERIVED SILICA

Rice, a member of Gramineae family, usually has 20% to 22% of its production as rice husk (RH). So, RH is the prime byproduct generated during rice production. Maximum silica (SiO_2) accumulation in rice is found in its husk that fluctuates between 8.7 to 12.1% and averaging almost 10.6% [42]. Azat S. [43] obtained 99.66% pure silica from rice husk waste through green method. Sankar S. [44] synthesized silica nano particles from different varieties of RH through chemical treatment. In another work, Sankar S. et. al. [45] produced silica via sonochemical method. Costa JAS et. al. [46] extracted amorphous silica (SiO2) from the rice husk ashes (RHAs) obtained by inorganic acid treatment of two varieties. They found that acid treatment increase the silica purity by decreasing oxides impurities. Jin Hyung Lee[47] prepared high purity silica from RH by removing metallic impurities through chemical treatment. Hydrated amorphous silica is found in RH, which is similar to that found in other plants in the biosphere [48]. Gu S. [49] reported a novel two-staged thermal method to synthesis nano silica and lower pyrolyzing temperature is more suitable for pure silica. They also observed that the CO_2 is preferable than N_2 as pyrolyzing media to give good quality silica production. In 2015, the global production of rice was approximately 749.8 million tons with 150 million tons of estimated rice husk production [50]. The world is experiencing a cumulative increase in rice production. Fig. 3 presents the total year wise milled rice production around the world [51]. Accordingly, the RH generation has also increased dramatically. Generally, rice husk is burnt or disposed of as waste, creating various environmental and health problems. However, attention has been paid to use rice husk for heat and electricity production [52]. Moreover, silica production from RH is considered to be an attractive utilization process. Silica is an important raw material widely used in industries such as molecular sieve, zeolite, catalysis, ceramics, electronics and polymer industries [53,54].

For silica extraction from RH, thermal and chemical treatments are performed and to do this, an important step is the thermo-chemical decomposition of rice husk. Rice husk ash (RHA) production from RH can be done by various thermal decomposition methods, where different researchers followed different ones. Table 3 shows the methods followed by various researchers for RHA production. Accordingly, Zhang et al. [55] produced RHA by performing small-scale pyrolysis of RH and washed RH in a thermo-gravimetric analyzer heated up to 700^{0}C with a heating rate of 20^{0}C per minute and N_2 flow rate of100mL per minute. After that, calcination is performed in a muffle furnace under an air flow at the heating rate of 20°C per minute at 600°C for 2 hours. On the other hand, Zhang et al. [56] followed a slightly different path. They pyrolyzed rice husk for RHA generation in an electrical heating tube furnace at different temperatures starting from 200^{0}C up to 800^{0}C at a heating rate of 10^{0}C/min and carrier gas flow rate of 300mL/min for 1 hour. Tyagi et al. [57] produced RHA by the incineration of rice husk in a muffle furnace at 650ºC. Again, Rhaman et al. [58] followed a pyrolysis technique for RHA production.

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They used a horizontal tube furnace made of 1.5 m long steel tube with 100 mm internal diameter for pyrolysis optimized temperature of 700^{0}C and controlled nitrogen gas flow. Hsu et al. [59] also used pyrolysis technique but different pyrolyzer and parameter. They pyrolyzed rice husk in a fluidized bed pyrolyzer using glass beads as the fluidizing media with the rice husk feeding rate of 10 g/min and the fluidizing nitrogen gas flow rate of 40 L/min. V.B. Carmona [60] successfully extracted and characterized nano and micro size silica by different acid solutions treatment. Khan et al. [61] used a special incinerator to produce RHA with a silica content of 97.4%. Moreover, RHA production by RH pyrolysis using flash pyrolysis method in a conical spouted bed reactor at 500^{0}C is utilized by Alvarez et al. [62].On the other hand, Zain et al. [63], Habeeb et al. [64] and Ramezanianpour et al. [65] used a different type of furnace with various combustion temperature and time as described in Table 3. Finally, Nehdi et al. [66] produced RHA by fluidized beds based on Torbed reactor technique. Better temperature control, economical and small RHA particle are the main features of this technique.

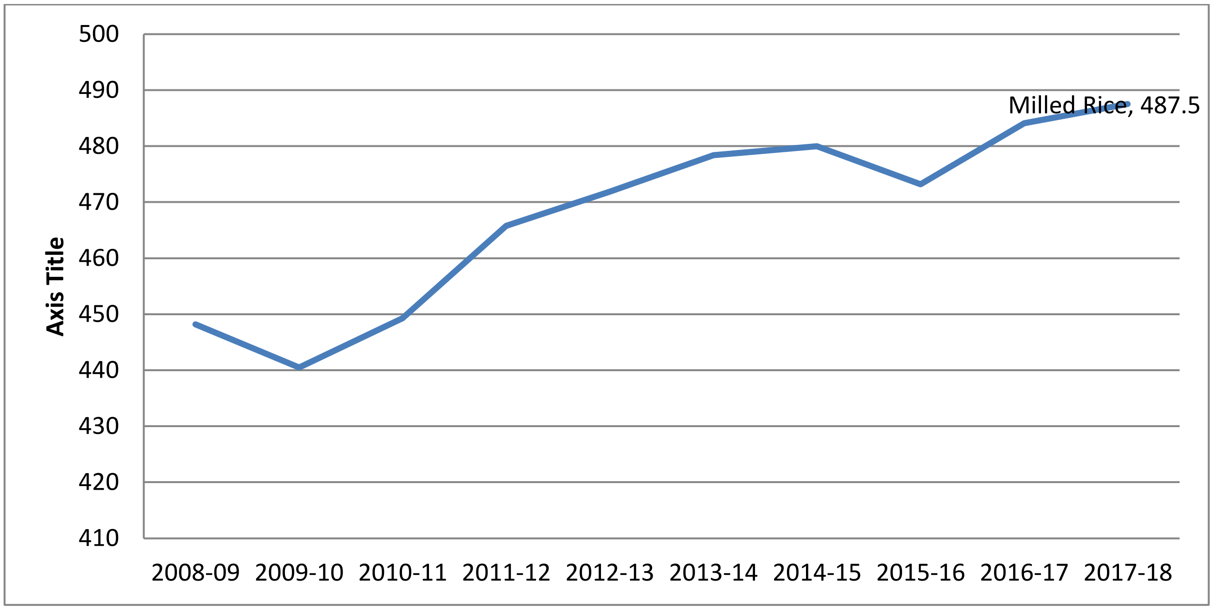

Fig. 3: Worlds yearly milled rice production in million metric tons

Table 3: Methods of ash production from rice husk found in different studies

Process Conditions	Reference
Small-scale Pyrolysis carried out in a thermo-gravimetric analyzer at 700 °C with the heating rate of 20°C/min and N_2 flow rate of 100 mL/min.	[55]
Pyrolysis in an electrical heating furnace at different temperatures ranging from 200^{0}C to 800^{0}C at a heating rate of 10^{0}C/min and carrier gas flow rate of 300mL/min.	[56]
Incinerating of husk at 650°C in a muffle furnace.	[57]
Pyrolysis at 700^{0}C and controlled nitrogen gas flow in a horizontal tube furnace made of 1.5 m long steel tube with 100 mm internal diameter.	[58]
Pyrolysis of Rice husk in a fluidized bed pyrolyzer using glass beads as the fluidizing media with a constant flow of nitrogen as the fluidizing gas.	[59]
Incineration of RH within a burning period of two days in one batch. The RHA produced by the method contains a large amount of amorphous silica.	[60]
Flash pyrolysis in a conical spouted bed reactor at 500^{0}C.	[61]

Ferrocement furnace combustion at 500^{0}C to700^{0}C for 2 h.	[62]
Incineration in a furnace with three small openings for ventilation with burning temperature of rice husk not more than 690^{o}C.	[63]
Furnace is used to produce RHA where burning temperature and rate of burning are easily measurable. Ash was produced at differenttemperatures such as 550, 600, 650, 700 and 750^{0}C with burning duration of 30, 60 and 90 mins respectively.	[64]
Use of fluidized beds based on Torbed reactor technique for RH burning. The Torbed reactor can operate with gas velocities of 3–12 m/s and it is easy to maintain temperature and economical.	[65]

V. REVIEW OF SILICA CONTENT IN RICE HUSK DERIVED ASH (RHA)

After the thermal decomposition of RH, RHA is obtained. This RHA is highly rich in silica. The silica content of RHA found in different studies is presented in Table 4. It shows that RHA has high SiO_2 content exciting 72%. It also indicates that ash derived from RH can be a cheap, renewable and natural source of SiO_2. According to Table 4, researchers found as high as 97.53% SiO_2 in RHA and the lowest SiO_2 content found is 73.60% [67,68]. In recent year studies, Zhang et al. found 91.98% SiO_2 in RHA by washing Pretreatment and first pyrolysis method in 2017[55]. On the other hand, Foong et al. in 2015 showed 93.46% SiO_2 content of RHA. Other recent year studies show RHA SiO_2 content less than 90%. Bakar RA [92] produced high purity silica from rice husk. During the production, they treated the rice husk at different temperatures after acid leaching. X-Ray Fluorescence (XRF) analysis confirmed maximum 99% pure silica from rice husk obtained at 600^{0}C for 2 hours. Chun J et. al. prepared high purity rice husk derived mesoporous silica with ordered porosity of different sizes [97].

Table 4: Percentage of silica in rice husk derived ash found by various researchers

SiO_2	Al_2O_3	Na_2O	CaO	K_2O	Fe_2O_3	MgO	SO_3	Reference
91.98	0.076	0.067	0.90	3.44	0.21	0.32	0.52	[55]
86.26	0.17	0.07	1.09	3.08	0.69	0.46	1.15	[56]
87.20	0.15	-	0.55	-	0.16	0.35	0.24	[69]
85.76	0.25	-	0.74	-	1.15	0.81	0.31	[70]
93.46	0.58	0.08	1.03	1.82	0.52	0.51	-	[71]
89.86	0.73	-	0.91	-	1.28	1.16	-	[72]
87.00	0.80	0.40	1.20	2.63	0.40	0.60	0.40	[73]
87.20	0.15	-	0.55	-	0.16	0.35	0.24	[74]
87.65	0.22	1.10	0.39	2.98	0.24	0.28	0.15	[75]
86.81	0.50	0.69	1.04	3.16	0.87	0.85	-	[76]
93.80	0.74	0.28	0.89	0.12	0.30	0.32	-	[77]
75.00	1.29	0.40	3.30	1.50	0.78	0.22	0.20	[78]
94.84	0.39	0.11	1.32	1.45	0.54	0.40	0.01	[79]
91.00	0.35	0.08	-	3.21	0.41	0.81	1.21	[80]
90.90	0.83	-	0.80	-	0.60	0.56	-	[81]
89.87	0.14	0.25	0.49	2.16	0.94	-	-	[82]
88.32	0.46	-	0.67	2.91	0.67	0.44	-	[64]
87.20	0.15	-	0.55	-	0.16	0.35	0.24	[84]
77.19	6.19	0.00	2.88	1.81	3.65	1.45	-	[85]
73.60	0.07	0.09	0.76	1.17	0.26	0.27	-	[68]
90.00	0.28	0.08	0.45	1.55	0.14	0.28	0.02	[86]
89.47	Traces	2.09	2.69	0.83	0.62	1.16	0.93	[87]
91.3	0.30	0.52	0.21	2.65	0.12	0.31	0.24	[88]

89.61	0.04	0.07	0.91	1.58	0.22	0.42	-	[65]
87.46	2.53	-	0.78	4.73	0.40	1.58	-	[89]
97.53	-	0.01	0.22	0.04	0.21	-	-	[67]
93.20	0.40	0.10	1.10	1.30	0.10	0.10	0.90	[33]
87.20	0.15	1.12	0.55	3.68	0.16	0.35	0.24	[91]
87.20	0.15	1.12	0.55	3.60	0.16	0.35	-	[93]
86.98	0.84	0.11	1.40	2.46	0.73	0.57	-	[94]
87.30	0.15	1.12	0.55	3.68	0.16	0.35	0.24	[95]

VI. FACTORS INFLUENCING CONTENT AND STRUCTURE OF SILICA IN RHA

Characteristics and properties of a product have a closerelationship with the properties of the parent raw material and the technique or method for the production. This is also applicable to RHA. Generally, RHA is generated from thermal degradation of rice husk through either open field burning or pyrolysis or controlled incineration. Due to pollution problems and poor RHA quality open burning of RH is not encouraged. The RHA generated by open burning usually has higher carbon content and highly crystalline structure with lower reactivity, which is generally not desired [96]. On the other hand, RHA with high SiO_2 content can be produced by pyrolysis technique [55,83,90].

6.1 Combustion method

Though RHA is mainly silica, the SiO_2 content of RHA found in different studies shows fluctuation between 70% and 99%. There are several reasons behind having difference in silica content. The major causes are various combustion techniques presented in Table 3 and chemical treatments that affect the SiO_2 content in RHA. Accordingly, Zhang et al. found 91.98% SiO_2 in RHA produced by pyrolysis method. Again, they were successful to obtained 99.33% SiO_2 in RHA by washing pretreatment of RH followed by pyrolysis [55]. Other researchers used various methods such as muffle furnace incineration, ferrocement furnace combustion, electrical heating furnace pyrolysis, fluidized bed pyrolysis or even open air combustion. Among these methods, washing pretreatment followed by pyrolysis showed better results [55,58].

6.2 Geographical location

Another reason is the geographic location of the paddy cultivation which affects the silica accumulation in rice husk (RH). So, ash derived from RH collected from different geographic locations has different SiO_2 content. Table 5 shows the SiO_2 content of RHA from different countries. Accordingly, researchers from different countries found different SiO_2 content in the local RHA, which indicates the effect of land and environment in the SiO_2 content of RHA. Moreover, silica accumulation in plants is basically a phylogenetic feature. Plant-available Si in the soil affects the silicon absorption of the plants[22,98].With intensive and continuous harvesting of silicon-accumulator crops like rice, the amount of plant-available silicon in the soil reduces significantly [99]. For example, if the rice productions in a field depend only on silicon present in the soil, the reserve of plant-available Si will be exhausted after five years of cultivation[100].In contrast, application of silicon fertilizer increases the Si content of the soil. These also affect the accumulation of Si in RH, causing the variation in SiO_2 content in RHA.

Table 5: Effect of soil and environment on silica and other mineral content in Ash

Countries	Percentage of minerals								Ref
	SiO_2	Al_2O_3	Na_2O	CaO	K_2O	Fe_2O_3	MgO	SO_3	
Bangladesh	89.86-91.30	0.30-0.73	1.52	0.21-0.91	2.65	0.12-1.28	0.31-1.16	0.24	[72] [88]
Brazil	92.9	0.18	0.02	1.03	0.72	0.43	0.35	0.10	[102]
Canada	87-97	0.15-0.4	0.10-1.12	0.40-0.49	2.0-3.0	0.16-0.4	0.35-0.50	0.0-0.24	[101]
Guyana	88-95	-	0.0-0.3	0.06-1.20	0.60-2.50	-	0.17-0.26	-	[101]

India	90.99	2.90	0.10	0.89	0.56	0.30	0.96	0.20	[102]
Iraq	86.8	0.40	1.15	1.40	3.84	0.19	0.37	1.54	[101]
Japan	91.6	0.14	0.09	0.58	2.54	0.06	0.26	0.52	[101]
Malaysia	93.1	0.21	-	0.41	2.31	0.21	1.59	-	[103]
Netherlands	86.9	0.84	0.11	1.4	2.46	0.73	0.57	-	[102]
Nigeria	67-76	3.0-4.90	-	1.36-6.0	0.0-0.1	0.0-0.95	1.30-1.81	0.0-2.8	[101]
Thailand	89-95	0.5-1.0	0.03-0.80	1.0-1.3	2.4-2.5	2.5-2.8	0.18-0.28	-	[101]
U.S.A.	87-97	Traces	0.0-0.15	0.25-1.0	0.58-2.0	0.38-0.54	0.12-2.0	0.0-1.13	[101]
Vietnam	86.90	0.84	0.11	1.40	2.46	0.73	0.57	-	[101]

6.3 Combustion temperature

On the other hand, the combustion temperature affects not only the SiO_2content but also the structure of the RHA derived SiO_2. Generally, RH consists of cellulose (about 50%), lignin (25% to 30%) and silica (15% to 20%). During combustion, cellulose and lignin are burnt out leaving silica-rich ash and the burning temperature affects mainly SiO_2structure in the ash and has little effect on silica content. Table 6 presents the consequence of temperature on the RHA silica. Accordingly, the structure of RHA silica shifts from amorphous to the crystalline structure with temperature rise. In the temperatures ranging from 600ºC to 700ºC amorphous structure with the highest pore diameter is observed and after this temperature range, crystal formation is experienced [103-105]. The difference in properties has been observed in amorphous and crystalline silica. So, it is crucial to determine the operating temperature during pyrolysis or incineration to get RHA with the desired structure.

Table 6: Effect of temperature on the structure of RHA silica [103-105]

Temperature,ºC	Description of the structure	Crystallinity
Within 500	Porous structure with spherical or globular particles	Amorphous
500 - 600	Non-crystalline and partially crystalline structure with the presence of fine porous crystalline grains. Reflecting the transmutation between the amorphous and the crystalline state	Amorphous
600 - 700	Amorphous particles with pores having the highest pore diameter	Amorphous
700 - 800	Relatively crystalline structure, exhibiting formation of coral-shaped crystals	Mixed
800 - 900	Crystalline structure	Crystalline
900 - 1000	Intensification in the formation of coral-shaped crystals	Crystalline

VII. METHODS OF SILICA EXTRACTION

Silica is a prime raw material extensively used in industries like molecular sieve, zeolite, semiconductor, ceramic and polymer industries [106]. Silica is produced from quartz that is fused at high temperatures [107]. This method has environmental and economic drawbacks. Due to advantages like low energy input and simpler methods of extraction, pure silica extraction from RH or RH derived ash can be a major solution for cheap and environment-friendly silica [108,109].

Different researchers optimized various methods to produce pure silica from RH or RHA. Table 7 presents a list of methods and their results utilized by different researchers. Accordingly, Zhang et al. [55] washed RH with aqueous fraction bio-oil and pyrolyzed at 700ºC. After that, calcination was done at 600°C for 2 hours in a muffle furnace and they obtained 99.33% pure amorphous silica. On the other hand, Tyagi et al. [57] acid treated RHA and then dispersed in NaOH and boiled for 1 h. The solutions were filtered, rinsed with boiling water, titrated against HCl, aged for at least 12 hours, crushed, washed and finally dried. The

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researchers were able to produce amorphous silica that can be used in food applications. RH combustion, acid leaching of ash and then incineration at 700ºC in atmospheric method conditions to get spherical, completely amorphous silica particles was suggested by Sankar et al. [110]. Bakar et al. washed RH with distilled water, acid leached and finally incinerated at different temperatures. They produced 99.58% pure amorphous silica particles by HCl leaching and incineration at 600ºC [111]. Rhaman et al. [58] followed a slightly different technique. They washed RH with distilled water, pyrolyzed in a horizontal tube furnace and finally extracted silica by alkali (NaOH) extraction method. They synthesized Porous and multifaceted amorphous silica. Alvarez et al. [62] used flash pyrolysis and chemical treatment with HCl and Na_2CO_3 solution to obtain amorphous silica and activated carbon products. Ma et al. [112] obtained Spherical, completely amorphous silica particles by chemical treatment of RHA with acid and NH_4F. Liou et al. [113] were able to obtain 99.48% purity silica by HCl leaching, incineration and extraction with NaOH. Fernandes [114] utilized an almost similar method to extract silica from RHA except for the use of H_2SO_4.Yalçin et al. [115] extracted 99.66% amorphous silica by water washing of RH, acid leaching, alkaline leachingand then Incineration at 600ºC. Finally, Kalapathy et al. [116] observed high silica yield byacid leaching treatment, SiO_2 extraction with NaOH, precipitation with HCl and then water washing.

Table 7: Methods of silica synthesis from RH and RHA

Method summary	Results	Reference
1) Washing pretreatment of RH with aqueous fraction bio-oil 2)Pyrolysis at 700ºC 3)Calcination in a muffle furnace at 600 °C for 2 h	99.33% pure amorphous silica was obtained.	[55]
1) Acid pretreatment of RHA 2) Dispersion in NaOH and boiled for 1 h 3) Filtered and titrated against HCl 4) Aged and crushed.	Amorphous silica that can retard fungal growth and can be used in food applications.	[51]
1) Combustion of RH. 2) Acid leaching 3) Incineration at 700ºC in atmospheric conditions	Spherical and amorphous silica particles with large specific surface area.	[110]
1) Washing of RH with distilled water 2) Acid leaching 3) Incineration at 500, 600, 700, 800, and 900 ºC for 2 h in atmospheric Conditions	Amorphous silica particles and HCl leached RH showed highest silica content of 99.58% at 600 ºC.	[111]
1)Washing of RH with distilled water and drying 2) Pyrolysis in a horizontal tube furnace 3) alkali (NaOH) extraction	Porous and multifaceted amorphous silica was synthesized.	[58]
1)HCl acid pretreatment 2) Silica removal with Na_2CO_3 solution 3) Carbonization for silica recovery	Amorphous silica and activated carbon production with high (88%) silica recovery.	[62]
1) Acid leaching of RHA 2) Extraction with NH_4F 3) Acid precipitation of SiO_2	Spherical and amorphous silica particles with 50 to 60 nm diameter and 94.6% yields observed.	[112]
1) Leaching of RH with water and then HCl 2) Incineration 3) Extraction with NaOH	99.48% purity silica at 50ºC, aging time of 12 h and pH 3.	[113]
1) Extraction of silica with NaOH	Colloidal silica with high reactivity	[114]

2) Precipitation using H_2SO_4 3) Filtration	was obtained and energy input was low.	
1) Washing of RH with water 2) Acid leaching 3) Alkaline leaching 4) Incineration at 600ºC under various conditions	Amorphous silica with a specific surface area of 321 m2/g and purity of 99.66% was obtained.	[115]
1) Acid leaching treatment 2) Extraction of SiO_2 with NaOH 3) Precipitation with HCl then water washing	Silica yield was magnificent (extraction with 1N NaOH). Washing with water lowered Na and K content.	[116]

Silica-rich RHA can be obtained by thermal treatment of RH and silica can be extracted from RHA in the form of sodium silicate by solvent extraction method. However, there are some other methods for silica extraction from RH or RHA. Fig. 4 presents the available methods of silica extraction from RH [55,117-127]. Considering the above-mentioned synthesis methods, the chemical methods with washing pretreatment and post heat treatment are considered as the most simple and successful SiO_2 extraction methods from RH[128]. Moreover, the properties and structure of the synthesized silica are highly dependent on the methods, temperature and chemical treatments that are utilized. The results may differ from amorphous to distinct crystal silica particles. Thus, RH can be considered as a significant source of silica[129].

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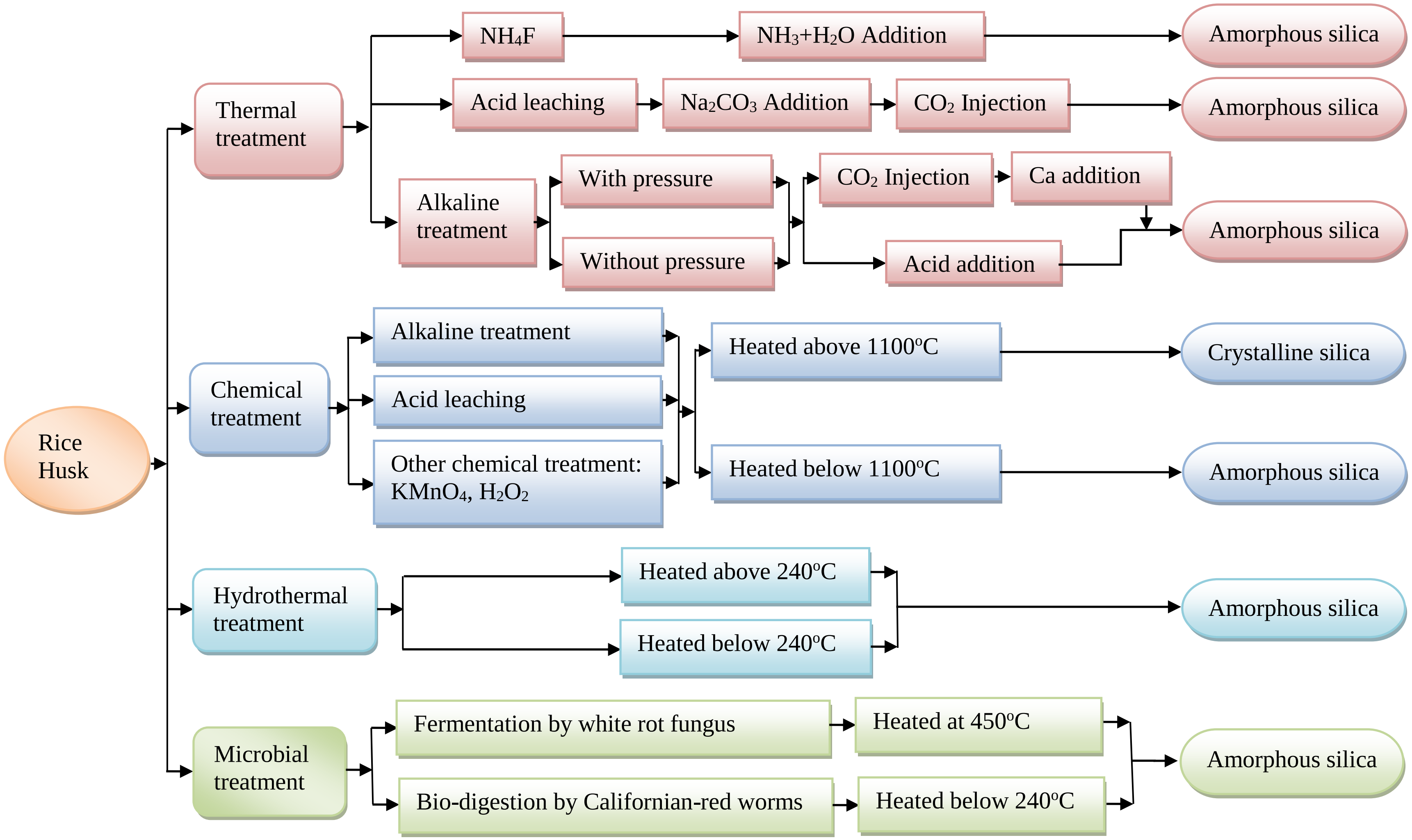

Fig. 4. Different silica extraction method from RH

VIII. APPLICATION OF RH DERIVED SILICA

Jamal D. [132] successfully synthesised 3,4-dihydropyrimidinones/ thiones compounds using RH derived nano-silica. Enrique C. Peres [133] obtained bio-silica from RH using ultra sound treatment and used as efficient dye adsorbent. Li W [134] proposed RH derived silica as an excellent candidate to design optoelectronic materials.

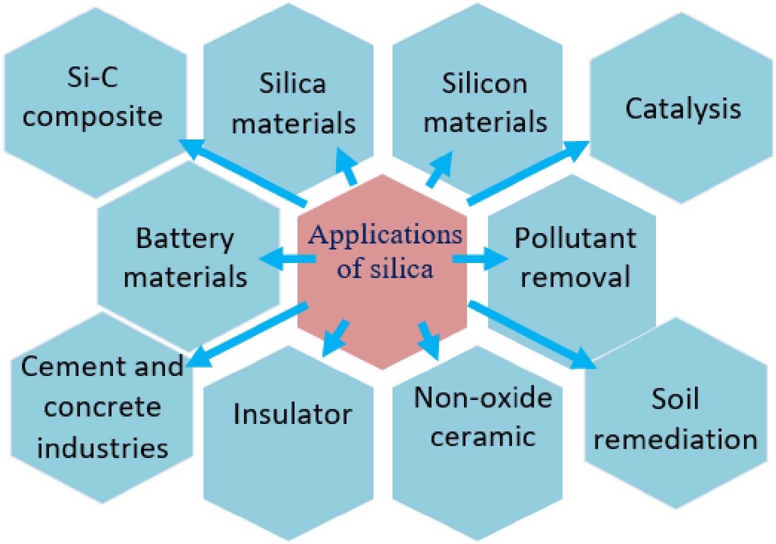

Fig. 5: Excellent applications of rice husk derived silica [shen2014, shen2017]

Mostafa Y. N [135] designed an economical and simple approach to fabricate tunable pure nanostructured silica that exhibited excellent adsorption of ciprofloxacin drug from aqueous solutions. Yusoff NM developed flowable composite using RH derived silica [136]. Belite cement was synthesized by using silica with low activation energy of dehydration and high pore volume from RH ash [137]. Sobrosa F.Z. [138] replaced kaolin to develop refractory ceramics using silica derived from RH ash. Wang Y. [139] synthesized ZSM-5 zeolite which showed excellent carbondioxide (CO_2) adsorption properties.

IX. MOLECULAR SIEVE

Molecular sieves are alkali metal aluminosilicate compounds with defined pore and characteristic geometrical shape that allow selective gas to pass from gaseous mixture according to their molecular size[140]. Pore size and pore size distribution are the key features to be characterized well to optimize its performance. A good quality molecular sieve can be synthesized by optimizing synthesis process, temperature, mixing ratio and reaction time [141, 142, 143]. It is not easy to define and categories zeolites because of its complex crystallinity [144, 145]. Zeolites have primary and secondary building units that are attached with each other to form defined cage (figure) [146].

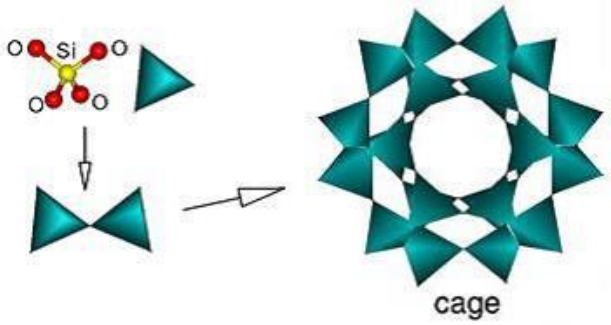

Fig. 6: Formation of cage combining PBUs and SBUs [146]

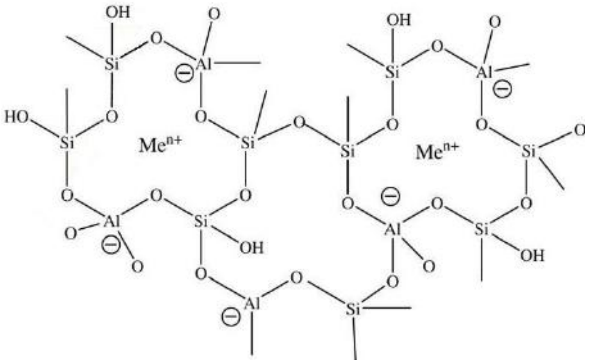

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Fig. 7: A two-dimensional framework of zeolites molecular sieve [147, 148]

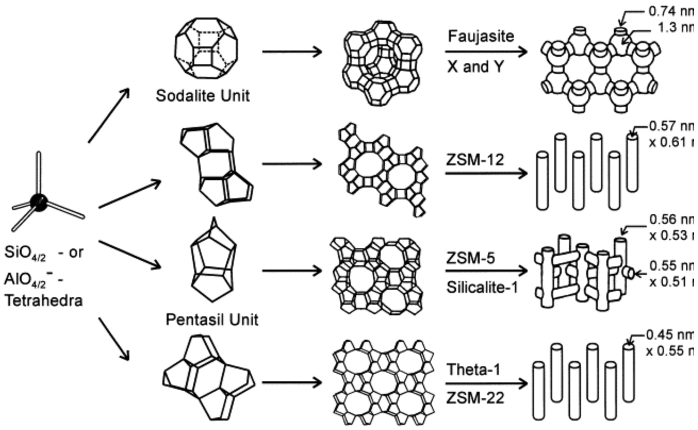

Fig. 8: Crystallographic structures of zeolite molecular sieve [146]

Generally, CBUs do not show chirality and do not form the total structure like SBUs [149]. When a ring is consisted of n number of tetrahedrons, it is called n-ring. Here n may be 4, 5, 6, 8, 10, or 12 in numbers and sometime it is 14, 18 or 20 [150-154], though 3-, 7-, or 9-ring structures are very rare [155-157].

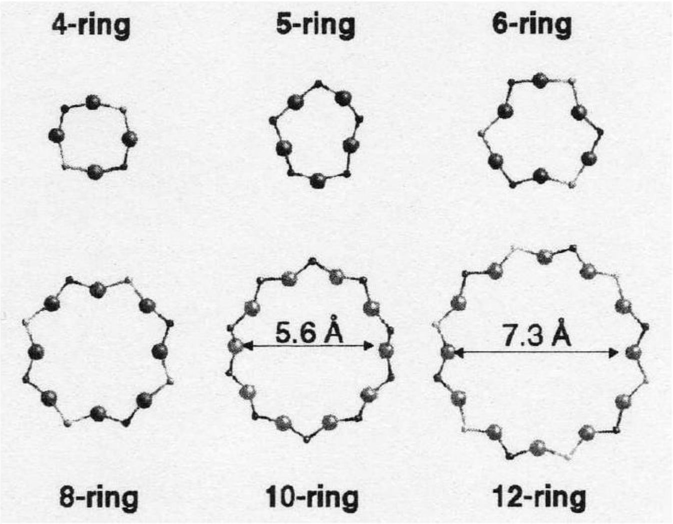

Fig. 9: Zeolite framework with different composition and size of rings

From the Fig. 9, the pore size of different zeolites is determined by the rings present in the corresponding framework [158]. The entrance to the pore is known as a window [159].

9.1 Properties & applications of zeolite molecular sieve

Zeolites as molecular sieve have drawn much attention due to its multidimensional properties. Ion exchange, catalytic, defined porosity and adsorption are key characteristics of zeolite molecular sieve. Some selective applications of zeolites should be mentioned, such as chemical sensors [160], separation of straight-chain hydrocarbons from branched-chain hydrocarbons [161], effluent and auto-exhaust control, industrial process control, environmental and indoor air-quality monitoring, medical monitoring [162], removal of heavy metals [163] and air separation [164]. Key applications are stated in following section.

Table 8: Application and properties of zeolite molecular sieve

Application of zeolite molecular sieve	Specific properties
a) Petrochemical refinery (Hydrocracking, catalytic cracking, hydrotreating, the Fischer–Tropsch process, alkylation, etc.)	Catalytic, high acidity, thermal stability and molecular sieve properties
b) Purification of Water (Industrial Wastewater, Municipal Wastewater, Drinking Water)	Cation Exchange Capacity [170], Adsorption Kinetics [171,172,173], Adsorption Isotherms [172,174,175] - Zeolites shows excellent adsorption of H_2O, H_2S,NH_3, NO, SO_2, NO_2, and CO_2 [176]
c) Formation of Sludge	pH dependent adsorption [177],

d) Medicinal Uses of Zeolites Treatment of Cancer,	mechanically activated clinoptilolite zeolites [178]
e) Catalysis Fine chemicals, detergents, dyestuffs, and scents can be produced by using zeolite as catalysts [9,179,180].	Proton Transfer, Zeolite Pore Size, Shape Selective Catalysis, Ion Exchange

9.2 Synthesis

Numerous articles demonstrating the methods of synthesizing molecular sieves using RH as source of silica are available [165-169]. Following is the short description of preparing different molecular sieves. Mainly, three types of zeolite preparation methods are:

i. Hydrothermal, ii. Solvo-thermal, iii. Iono-thermal [181]

In *hydrothermal* process, water is used as mineralizer keeping the temperature range at 90-180^{0}C and Pressure maximum 15 bars during synthesizing molecular sieve in this process. Low operating temperature makes this method easy and cheap. Molecular sieve with completely new properties were produced at high temperature (above 200^{0}C) and pressure (2000bar) [182].

Zhang P [183] proposed a two-stage hydrothermal process to produce Silicoaluminophosphate (SAPO)-11 that shows excellent hydroisomerization catalytical properties.

X. MOLECULAR SIEVES SYNTHESIS USING RH DERIVED SILICA

Molecular sieves were synthesized from commercial chemicals but it is expensive and environmentally hazardous. Qian J [184] reported the successful preparation of nano size zeolitic imidazolate framework-67 through hydrothermal method. In search of alternatives, different industrial and geological sources are being discovered over time.

Table 9: Syntheses of molecular sieve from RH derived silica with different composition

Method	Feed	Si in RHA (%)	Molecular sieve features	Ref
Hydrothermal	Only rice husk ash	95.91	ZSM-5	[181]
Hydrothermal	RH & RHA	95.62	MCM-41	[182]
Hydrothermal	Glycolato silicate and synthetic zeolite(NaY)	98.30	Maximum 98.76% product yield, SiO_2:Al_2O_3=5.4	[184]
Hydrothermal	RHA derived Al_2O_3:$10SiO_2$ as seed gel	89.00	In 1 step: less pure In 2 step: purer	[185]
Hydrothermal	Al_2O_3 as Al source and RH silica	81.60	68% crystal (33nm), SiO_2/Al_2O_3 = 1.25	[186]
Hydrothermal	Composite RHA	--	4A type, 85% crystalline	[187]
Hydrothermal	Commercial chemical as seed and RHA as feedstock,	92.00	NaY type	[188]
Hydrothermal	Sodium aluminate and RHA derived silica	99.64	ZSM-5	[189]
Hydrothermal	Aluminum metal foil RHA derived silica	95.54	ZSM-5	[190]
Hydrothermal	Aluminum metal foil RHA derived silica	95.54	ZSM-11	[191]
Hydrothermal	Aluminum metal foil and RHA derived silica	95.54	Coral-like Hydroxysodalite	[192]

Hydrothermal	Sodium aluminate and RHA derived silica	91.65	NaA-type	[193]
Hydrothermal	RH derived silica and $NaAlO_2$	98.50	MCM-22	[194]
Microwave-hydrothermal	Molar composition = $20SiO_2$ (in the carbonized rice husk):10NaOH: 1TPABr:$2800H_2O$	91.69	ZSM-5	[195]

Rice husk ash Table 8, Coal fly ash [185], high silica fly ash [186], waste sand stone cake [187,188], bagasse fly ash [189], perlite [190], agricultural waste [191], halloysite paper sludge [192] etc. are the commonly used as sources of silica which is one of the constitutional compound of molecular sieves. 89.56% crystalline ZSM-5 Molecular sieve was synthesized successfully through hydrothermal process without using any alumina source. They used RHA with 95.91% silica and tetrapropylammonium bromide (TPABr) and tetrapropylammonium hydroxide (TPAOH)) as template reagents to support the formation of the molecular sieve framework. Different alumina sources were also taken to prepare samples for comparative study [193]. MCM-41 type was synthesized with RH and RHA of 95.62% silica source and initial mixing ratio was $1.0SiO_2$:0.13CTAB:1.1NaOH:$0.12H_2O$ at pH 10. Trimethylchlorosilane (TMCS) and phenyldimethylchlorosilane (PDMS) were used as silylating agent. The purpose of the study was to understand the effect of further processing RH to RHA on hydrophobicity. RH-MCM-41 showed higher hydrophobic property at the presence of silylating agents [194]. Highly crystal molecular sieves were prepared by glycolato silicate and synthetic NaY (formula: $Na_{32}(AlO_2)_{32}(SiO_2)_{176}192H_2O$) with RH derived ash with silica content 98.30% and found the 98.76% product yield of SiO_2/Al_2O_3 ratio 5.4. The stability of the product was tested with TGA-DTA and dehydration occurred at temperature range 300^0C-500^0C. They observed that product crystallinity was not obtained before Si/Al ratio to 4.84 [195].

Alyosef H [196] reported the formation MFI type zeolite molecular sieve from rice husk ash at 175^0C temperature in different crystallization times and steps. They found a good quality zeolite after the four day crystallization and two steps method. Wang Y [197] prepared three types of zeolite molecular sieve through improved hydrothermal method and investigated their performances. Modified hydrothermal method showed superior potentiality over traditional hydrothermal method. Mohamed R [198] prepared molecular sieve using feedstock gel and seed gel. Total solution was divided into two parts, one part was allowed to be crystallized at 100^0C temperature for 24h after being aged at ambient temperature for 24h and another part was crystallized in autoclave at 110^0C temperature for 24h. Salama T [199] used different ratio of feed materials. They maintained molar fraction of the reaction gel was: xNa_2O: 1.76 Al_2O_3: $ySiO_2$: $125H_2O$, where x/y ratio was between 0.69 and 1.01. 80^0C to 100^0C was chosen for crystallization temperature for the period of 2 & 4 days and pH was 11 of another gel composition: $2.76Na_2O$:$1.76Al_2O_3$:$2.5SiO_2$:$125H_2O$. Mane S [200] used a composite made with coal ash and RHA as source of silica to evaluate the performance of different silica sources for 4A type molecular sieve preparation. Crystallization time was 3h and temperature 98-100^0C, finally experimental results confirmed the formation of solid crystal molecular sieve similar to 4A. The product quality resembled that of commercial chemicals derived molecular sieve. Tan W-C [201] produced molecular sieve using 92% pure silica and investigated the influence of commercially used silica and RH derived silica on the properties of NaY and NaA type molecular sieve. Panpa W. [202] prepared ZSM-5 molecular sieve with a wide range of Si/Al ratio at temperature 150^0C, pH 11 and crystallization time 4h-24h. Effect of crystallization time, temperature and Si/Al ratio was successfully experimented. Naskar MK [203] prepared ZSM-5 molecular sieve from aluminum foil as alumina source. RHA calcined at 700^0C for 6h, having 95.54% silica was used as silica source. Initial molar fraction was Na_2O:100-SiO_2:Al_2O_3:20TPAOH:$4000H_2O$ at operation temperature from 130^0C to 170^0C. They obtained improved surface area, fine particle size and well crystalline structure. Dey KP [109] got ZSM-11 molecular sieve from crystallization between aluminum metal foil derived aluminum solution as Aluminum source and RHA with 95.54 as silica source at 100^0C for 12days, molar fraction $4Na_2O$:$100SiO_2$:Al_2O_3:35TBAOH:$1200H_2O$. Same Si and Al sources are used by Naskar MK [108] to Synthesize Coral-like Hydroxy sodalite, formula:$[Na_8[Al(SiO_4)]6Cl_2]$. Yusof AM [204] prepared molecular sieve faujasite-type with primary molar fraction of 6Na2O:Al2O3:5SiO2:315H2O at 100^0C for 7-8h and NaA with primary molar fraction $3.165Na_2O:Al_2O_3:1.926SiO_2:128H_2O$ at 100^0C for 6h from RHA of above 90% silica purity and studied the effect of seeding and ageing technique. Final product was metastable and found no impurities. Cheng Y [205] reported the crystallization time reduction from 9-12days to 3 days as the influence of varying-temperature conditions in MCM-22 preparation. Smaller size ZSM-5 was prepared as the result of microwave radiation during dissolution of silica through hydrothermal process Katsuki H [206]. In *solvothermal* different solvents are used in this process. By definition, it also can be classified as hydrothermal method mentioned above. The choice of solvent depends on polar-hydrophobic and nonpolar-hydrophilic nature. Alcohol, pyridine and different hydrocarbons are commonly used solvents [182]. In *ionothermal* ionic compounds get preference other than solvents used in the solvothermal method and these ions are commonly known as ionic liquids, unlike molecular nature. Therefore, ionic liquids properties vary with typical properties like low vapor pressure [182]. Peres EC [211] synthesized molecular sieve using ultrasound method and observed its excellent performance.

XI. PARAMETERS AFFECTING THE SYNTHESIS OF MOLECULAR SIEVES USING RH AS SILICA SOURCE

Different parameters are maintained during synthesis of molecular sieves from rice husk derived silica. Molar ratio of mixing ingredients, reaction conditions [199], like temperature, reaction time, acid-base media, ageing etc. are studied over the time.

11.1 Si/Al ratio

The properties and types of MS are significantly determined by silicon/aluminum molar ratio [33, 36] Table 9. Panpa W [202] studied detail effect of the Si/Al ratio from 30 to 2075 on the characteristics of MS formed by hydrothermal one and two step processes. As Panpa W reported that the BET surface area increased to maximum 365 with the increment of the Si/Al till 200 and then that decreased for further increment of Si/Al ratio. Amorphous MS was observed till 50 [fig. 7(a)] and crystallinity started from Si/Al ratio 80 while Silicalite product was formed after ratio 200 Table 10.

Table 10: Si/Al effect on BET surface area and crystal formation of MS

Si/Al ratio	BET surface area (m^2/g)	MS phase formed	Total pore volume (cm^3/g)	Ref
30	27	Amorphous	0.0358	**[189]**
50	82	Amorphous	0.0605	**[189]**
80	306	ZSM-5	0.1454	**[189]**
100	308	ZSM-5	0.1758	**[189]**
200	365	ZSM-5	0.1871	**[189]**
400	315	Silicalite	0.1384	**[189]**
40	--	No MS crystal	--	**[168]**
176	--	ZSM-5	--	**[168]**

SEM image showing the morphology of MS with Si/Al molar ratio 200 revealed the finely formation of crystallized particles fig. 7. XRD of MS crystal confirmed that the higher Si/Al ratio increased the crystallinity of molecular sieves and well defined crystal was formed for Si/Al ratio 200(Fig. 10).

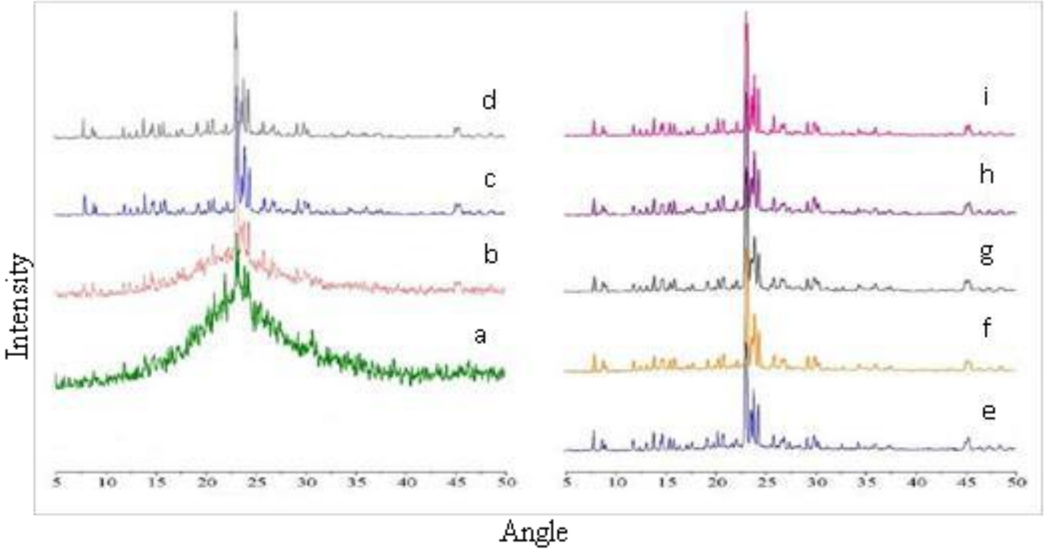

Fig. 10: XRD pattern of Molecular Sieves of Si/Al ratios of 30(a), 50(b), 80(c), 100(d), 200(e,) 400(f), 800(g), 1000(h), 2075(i)

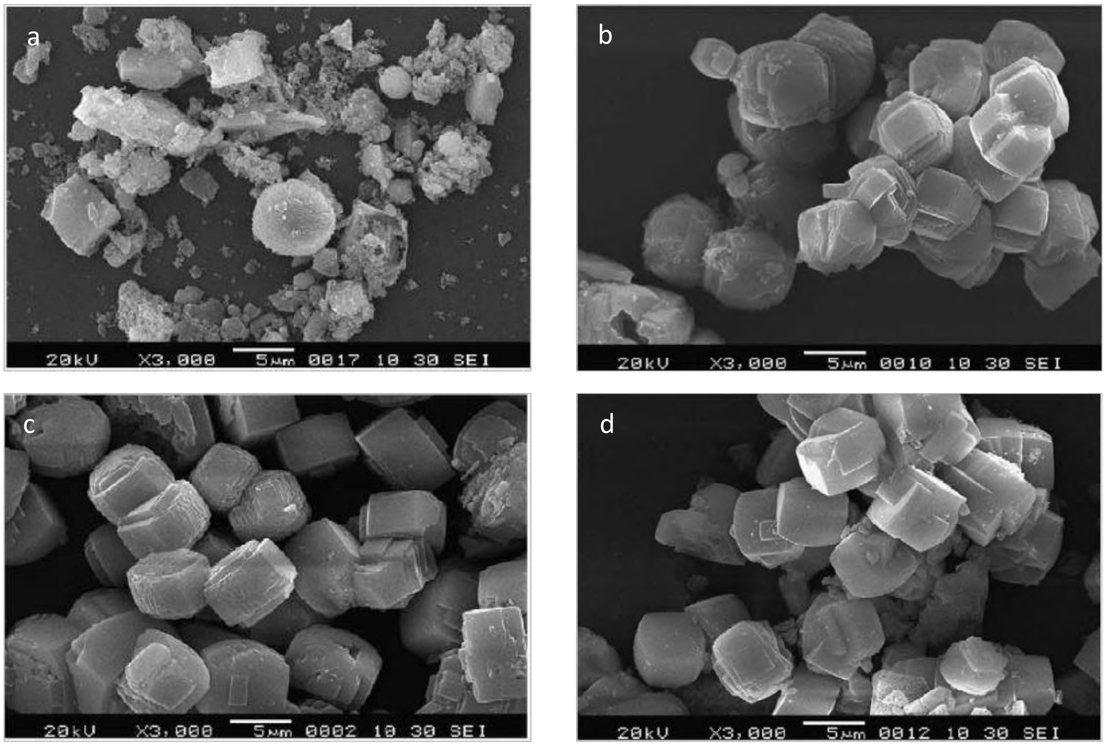

Fig. 11: SEM image of MS with Si/Al ratio 50(a), 100(b), 200(c), 400(d)[189]

Z.G.L.V. Sari [137] concluded that no ZSM-5 crystal was found in case of Si/Al ratio 40 instead of 176 at 150^{0}C. The rate of ZSM-5 formation increases with Si/Al ratio increment as reported by previous studies [207,208].

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Different morphologies and crystal structure are observed due to the ratio of Si and Al. Some images are given below with due permission of authors.

11.2 Crystallization time

Crystallization time is another important parameter that determines the final product quality and types. Some authors suggested that an optimum time which should be maintained to obtain certain types of MS with desired quality [189,194,203,209,210]. Mohamed R [166] prepared NaY type MS for 24h, after 24h duration NaP type MS started to form.

No peak in XRD results of NaY type MS was found for crystallization time 36 & 48 hours. Table 3 demonstrates the change of BET surface area and pore volume for different crystallization time. Maximum BET surface area (716.6 m^2/g) and pore volume (0.350cm^3/g) were detected by them [194] Table 11.

Table 11: Effect of crystallization time during MS synthesis

Time (h)	BET surface area (m^2/g)	Pore volume (cm^3/g)	MS Crystal	Ref
12	528.5	0.240	--	[182]
24	716.6	0.350	--	
36	32	0.006	--	
48	18.6	0.002	--	
48	--	--	No ZSM-5	[168]
96	--	--	Pure ZSM-5	[168]
7	--	--	Pure NaY	[190]
8	--	--	NaY with impurities	[190]

Z.G.L.V. Sari [193] did not find any ZSM-5 MS for 48 hours of reaction time and pure form of ZSM-5 crystal (Fig. 8.) was confirmed by XRD result obtain for 96 hours [fig. 12].

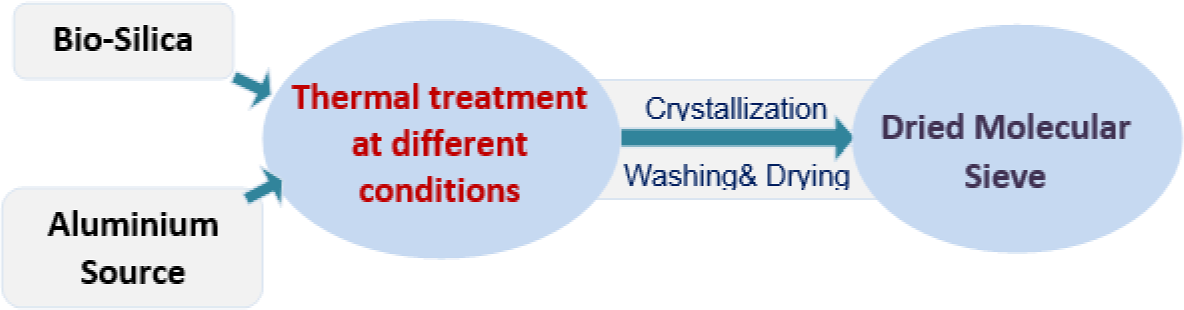

Fig. 12: Process diagram for Molecular sieves synthesis from RH derived silica.

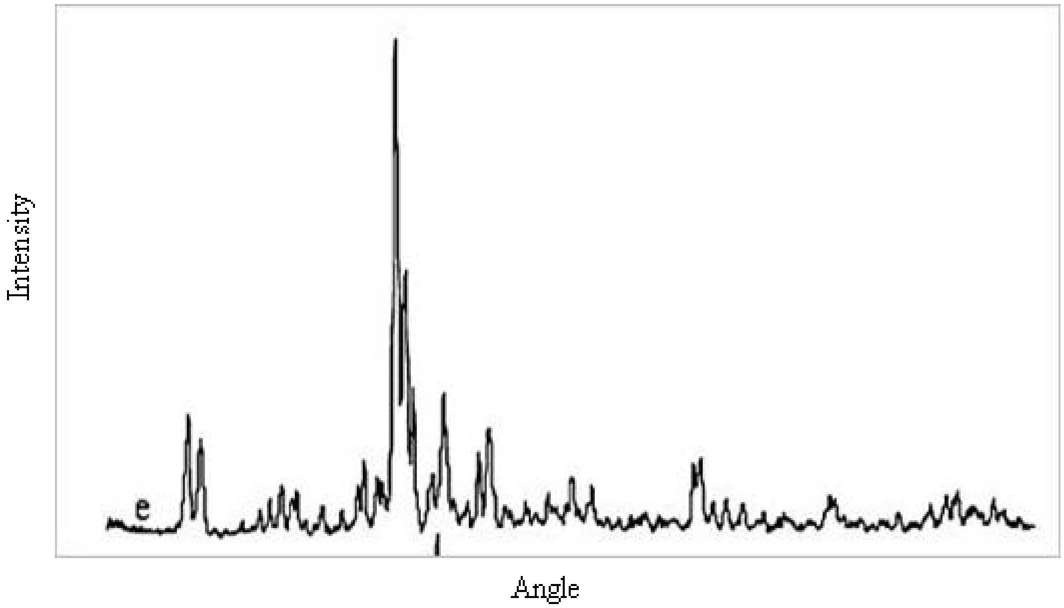

Fig. 13: XRD pattern of pure form of ZSM-5 molecular sieve

Yusof AM [204] observed that pure MS was found at 7hrs of crystallization while some impurities were presented 8hrs of crystallization that met the previous results. Bhavornthanayod C [209] studied the dependency of MS crystal formation on reaction time 4hr and 2hrs. They reported that 4hrs of reaction was more favorable than 2hrs. Rawtani AV [210] observed the rapid crystallization after 6hrs and 95% yield after 2.5days.

11.3 Ageing

Prepared solutions are aged for certain period of time before starting the crystallization. Ageing also contributes in modifying the properties of molecular sieves synthesized using RH derived silica. Increased pore volume and surface area were reported by some authors [212, 213]. Salama T [199] precisely studied the ageing time and temperature and suggested that four days ageing time give highest crystallinity and crystal product at 90^0C while amorphous product at 100^0C. Yusof AM [204] synthesized NaY type MS with ageing and without ageing and observed that with ageing ensured higher purity. Grisdanurak N [213] concentrated on the crystalline diameter influenced by different ageing time 144h, 96h, 48h, 24h. They concluded that crystal diameter reduced with ageing time increased Table 12.

Table 12: Relation between ageing time and crystal diameter

Ageing time(h)	**Mean diameter of crystal**
144	0.64
96	0.66
48	0.84
24	1.05

Mohamed R [198] aged the homogenous gel at room temperature for 24h before crystallization at 110^0C. XRD pattern of as synthesized MS was compared with that of standard Y type MS, almost similar pattern was revealed (in Fig. 14).

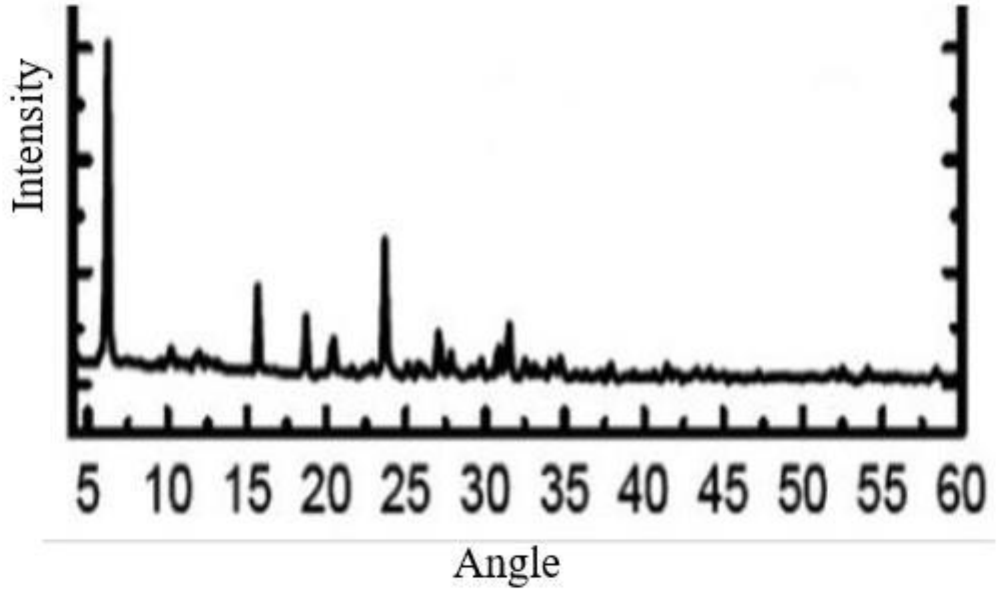

Fig. 14. Comparison between XRD pattern of standard Y type MS (a) and that of two step synthesized MS(b)

11.4 Temperature

Crystallization temperature is one of the most influential factors that determined the product formation rate, crystallinity etc. Bajpai PK [214] investigate the effect of temperatures (135^{0}C, 150^{0}C, 165^{0}C) on the formation of mordenite type molecular sieve using rice husk derive silica with the initial molar composition $3.5Na_2O.A1_2O_3.10SiO_2.219H_2O$ for 24h. No molecular Sieve was formed below 135 and amorphous product was in dominant for 135^{0}C and 150^{0}C temperature and crystal product for 165^{0}C. They also concluded that higher temperature is preferable for higher crystallization. *But* Cheng Y [215] found the greater amount of MCM-22 crystal at lower temperature. According to Sari ZGLV[193], ZSM-5 could not be produced at 150^{0}C while Chareonpanich M [216] observed the formation of ZSM-5 at 210^{0}C temperature at the same Si/Al ratio 40.

11.5 Others

Authors found optimum conditions for maximum good quality molecular Sieves yield. Crystallization reaction rate was increased using Microwave radiation as heat source [217]. Mane S [200] and Tan W-C [201] reported the composite silica source was able to alter the high cost and hazardous commercial chemical for molecular sieves fabrication Table 13.

Table 13: Miscellaneous Factors influencing molecular sieves synthesized using RH derived silica.

Parameters	Observations	Ref
Microwave Irradiation heat	Fast reaction	**[199]**
Composite silica source	Similar to commercial chemicals derived Molecular sieves	**[187]**
Silica source	Commercial silica can be replaced by RH derived silica	**[188]**
Effect of Starting Composition	Optimum molar portion of silica is 70%-80%	**[202]**
Effect of Alkalinity	Crystallinity decreases in basic media	**[202]**

Mixture seeding effects of precursors were investigated and findings told that high purity of product can be achieved by properly seeding [204]. According to Rawtani AV [210] optimum silica content in starting materials should be 70%-

80% and basic media for reaction was not suitable for crystallization process Table 12. Therefore, the review highlights some important uses or application of different types of molecular sieves and tabulated in table 14

Table 14: Uses of different types of molecular sieves

	Classification	Uses of different molecular sieves as classified	Remarks
Molecular sieves	13X, 1-2mm (0.04-0.08in) dia. pellets	Organic synthesis, pharmaceuticals and agrochemicals. It is preferred for static dehydration in closed liquid or gas systems, e.g., in packaging of drugs, electric components and perishable chemicals; water scavenging in printing and plastics systems and drying saturated hydrocarbon streams.	Hygroscopic. Ambient temperatures.
	13X, 1.6-2.5mm	They are used as catalyze carrier, co adsorption of CO_2 and H_2O and H_2O, H_2O and H_2S, as desiccant for medical and air compressor systems. Major applications in commercial gas drying, air plant feed purification (simultaneous H_2O and CO_2 removal) and liquid hydrocarbon/natural gas sweetening (H_2S and mercaptan removal).	
	13X, 3-4mm	commercial gas drying, air plant feed purification (simultaneous H_2O and CO_2 removal) and liquid hydrocarbon/natural gas sweetening (H2S and mercaptan removal).	
	13X, 3-5mm	used for liquefied petroleum gas, natural gas drying, desulfurization, clean. It is used as an adsorbent for gases and liquids.	
	3A, 0.4-0.8mm	used as adsorbents.	Hygroscopic. Store under Nitrogen. Ambient temperatures.
	3A, 1-2mm	used as a desiccant in petroleum and chemical industries, drying of unsaturated hydrocarbons such as ethylene, propylene and butadiene, removal of water from ethanol, drying of certain chemicals namely ethanol, refrigerants and natural gas. selective adsorption of water in insulated glass (IG) and polyurethane, drying agent in polar and non polar media.	Hygroscopic. Store under Nitrogen. Ambient temperatures.
	3A, 1-2mm (0.04-0.08in) dia. pellets	desiccant in petroleum and chemical industries, drying of unsaturated hydrocarbons such as ethylene, propylene and butadiene, removal of water from ethanol, drying of certain chemicals namely ethanol, refrigerants and natural gas, adsorption of water in insulated glass (IG) and polyurethane.	Hygroscopic. Store under Nitrogen. Ambient temperatures.
	3A, 2-5mm	Desiccant in petroleum and chemical industries, unsaturated hydrocarbons such as ethylene, propylene and butadiene, removal of water from ethanol, selective adsorption of water in insulated glass (IG) and polyurethane.	Hygroscopic. Store under Nitrogen. Ambient temperatures.
	3A, 3-4mm (0.12-0.16in) dia. pellets	desiccant in petroleum and chemical industries, drying of unsaturated hydrocarbons such as ethylene, propylene and butadiene, removal of water from ethanol, drying of certain chemicals namely ethanol, refrigerants and natural gas, selective adsorption of water in insulated glass (IG) and polyurethane.	Hygroscopic. Store under Nitrogen. Ambient temperatures.
	3A, powder	desiccant in petroleum and chemical industries, drying unsaturated hydrocarbons such as ethylene, propylene and butadiene, removal of	Hygroscopic. Store under Nitrogen.

		water from ethanol, drying of certain chemicals namely ethanol, refrigerants and natural gas	Ambient temperatures.
	4A, 0.4-0.8mm (0.02-0.03in) beads	general gas drying applications.	Hygroscopic. Ambient temperatures.
	4A, 1-2mm (0.04-0.08in) beads	absorb molecules as large as butane. separation of natural gas, liquified petroleum gas (LPG) and alkenes,refining and purification of liquid and gases, drying and purification of air, inert gases and solvents, remove methanol, carbon dioxide and ammonia from fluid streams, remove metal ions available in water	
	4A, 3-4mm (0.12-0.16in) dia. pellets	absorb molecules as large as butane. separation of natural gas, liquified petroleum gas (LPG) and alkenes, drying and purification of air, inert gases and solvents, involved to remove methanol, carbon dioxide and ammonia from fluid streams, remove heavy metal ions available in water, used as a soap forming agent and as a detergent auxiliary.	
	4A, 3-5mm (0.12-0.20in) beads	general gas drying applications.	
	4A, powder	absorb molecules as large as butane. separation of natural gas, liquified petroleum gas (LPG) and alkenes, refining and purification of liquid and gases, drying and purification of air, inert gases and solvents, remove methanol, carbon dioxide and ammonia from fluid streams, remove heavy metal ions available in water, soap forming agent and as a detergent auxiliary.	Hygroscopic. Ambient temperatures.
	4A with indicator, -8+12 (ca 2mm) beads	absorb molecules as large as butane. separation of natural gas, liquefied petroleum gas (LPG) and alkenes, refining and purification of liquid and gases, drying and purification of air, inert gases and solvents, remove methanol, carbon dioxide and ammonia from fluid stream, remove heavy metal ions available in water, soap forming agent, detergent auxiliary.	Hygroscopic. Store under Argon. Ambient temperatures.
	5A, 1-2mm (0.04-0.08in) dia. pellets	desiccation and purification, application in natural gas and liquid petroleum gas especially for dehydration and desulphurization, separating compounds and drying reaction starting materials.	
	5A, 3-4mm (0.12-0.16in) dia. pellets	as an sorbent.	
	5A, 3-5mm (0.12-0.20in) beads	For liquefied petroleum gas, natural gas drying, desulfurization, clean, adsorbent for gases and liquids, separating compounds and drying reaction starting materials.	
	tantalum oxide based; mesoporous,32 Angstrom, powder	liquefied petroleum gas, natural gas drying, desulfurization, adsorbent for gases and liquids, separating compounds and drying reaction starting materials.	
	titanium oxide based; mesoporous, 25 Angstrom, powder	oxidation and photo-catalysis	

XII. CONCLUSION

Materials design and engineering research play key role in supporting industry and to uplift the technologies. Many important applications or uses of molecular sieves drive extensive investigation further to achieve optimum performances of processes that bring breakthroughs. In-depth fundamental knowledge of physio-chemical properties and their modification techniques, incorporation methods in different devices like corrosion resistant, micro reactors, thermocouple, chiral frameworks, functional hierarchical materials, catalyst and adsorbent etc. direct to find economic and structural materials to meet the multidimensional demand. Therefore this review presented a complete path of molecular sieves production i.e; best feedstock, production technique, intermediate steps, required precautions, structural modification techniques, potential source/feedstock, factors affecting the production, structure and performances. Accordingly, RH can be an economical source of amorphous silica in Bangladesh and high quality MS can be synthesized utilizing this renewable bio-silica source.

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